Current and Future Linked Responses of Ozone and PM$_{2.5}$ to Emission Controls

KUO-JEN LIAO,[†] EFTHIMIOS TAGARIS,[†]
SERGEY L. NAPELENOK,[†,§]
KASEMSAN MANOMAIPHIBOON,[†,‖]
JUNG-HUN WOO,[‡,⊥] PRAVEEN AMAR,[‡]
SHAN HE,[‡] AND
ARMISTEAD G. RUSSELL*,[†]

School of Civil & Environmental Engineering, Georgia Institute of Technology, Atlanta, Georgia, Northeast States for Coordinated Air Use Management (NESCAUM), Boston, Massachusetts, Atmospheric Sciences Modeling Division, Air Resources Laboratory, National Oceanic and Atmospheric Administration, Research Triangle Park, North Carolina, Joint Graduate School of Energy and Environment, King Mongkut's University of Technology Thonburi, Bangkok, Thailand, and Department of Advanced Technology Fusion, Konkuk University, Seoul, Korea

Responses of ozone and PM$_{2.5}$ to emission changes are coupled because of interactions between their precursors. Here we show the interdependencies of ozone and PM$_{2.5}$ responses to emission changes in 2001 and 2050, with the future case accounting for both currently planned emission controls and climate change. Current responses of ozone and PM$_{2.5}$ to emissions are quantified and linked on a daily basis for five cities in the continental United States: Atlanta, Chicago, Houston, Los Angeles, and New York. Reductions in anthropogenic NO$_x$ emissions decrease 24-h average PM$_{2.5}$ levels but may either increase or decrease daily maximum 8-h average ozone levels. Regional ozone maxima for all the cities are more sensitive to NO$_x$ reductions than at the city center, particularly in New York and Chicago. Planned controls of anthropogenic NO$_x$ emissions lead to more positive responses to NO$_x$ reductions in the future. Sensitivities of ozone and PM$_{2.5}$ to anthropogenic VOC emissions are predicted to decrease between 2001 and 2050. Ammonium nitrate formation is predicted to be less ammonia-sensitive in 2050 than 2001 while the opposite is true for ammonium sulfate. Sensitivity of PM$_{2.5}$ to SO$_2$ and NO$_x$ emissions changes little between 2001 and 2050. Both ammonium sulfate and ammonium nitrate are predicted to decrease in sensitivity to SO$_2$ and NO$_x$ emissions between 2001 and 2050. The complexities, linkages, and daily changes in the pollutant responses to emission changes suggest that strategies developed to meet specific air quality standards should consider other air quality impacts as well.

---

* Corresponding author tel: 404-894-3079; e-mail: ted.russell@ce.gatech.edu.
[†] Georgia Institute of Technology.
[‡] Northeast States for Coordinated Air Use Management (NESCAUM).
[§] National Oceanic and Atmospheric Administration.
[‖] King Mongkut's University of Technology.
[⊥] Konkuk University.

Introduction

The formation of ground-level ozone and PM$_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5 μm) is strongly coupled because of their common sources, secondary nature, and interactions of their precursors (*1*). Changes in both climate and precursor emissions are expected to alter characteristics of ozone and secondary PM$_{2.5}$ (e.g., ammonium sulfate ((NH$_4$)$_2$SO$_4$), ammonium nitrate (NH$_4$NO$_3$), secondary organic aerosols (SOAs), etc.) formation and their interdependencies. Due to interactions between precursors of ozone and secondary PM$_{2.5}$, control measures for one pollutant may lead to increases in others, and reductions in one location may be accompanied by increases in others. For example, decreases in anthropogenic nitrogen oxides (NO$_x$) emissions reduce regional ozone maxima and PM$_{2.5}$ concentrations, but may increase concentrations of ground-level ozone in NO$_x$-rich areas. Likewise, reductions in sulfur dioxide (SO$_2$) emissions decrease sulfate levels but induce more nitrate formation (*2*). Unger et al. suggest that increases in emissions of ozone precursors will enhance sulfate formation up to 20% on a global scale in 2030 climate (*3*). For evaluating policy options, it is important to investigate the interdependencies between ozone and PM$_{2.5}$ formation and how those pollutants respond to emission controls currently and as conditions change in the future. Such information can be used to evaluate how controls developed for one purpose, e.g., meeting an air quality standard for one pollutant metric, might influence levels for other outcomes, e.g., overall health and welfare. Here we examine daily responses of ozone and PM$_{2.5}$ to emission changes for current and future scenarios, including effects of climate change and currently planned emission controls, and investigate their correlations.

Two frequently used indicators of air quality are the daily maximum 8-h average ozone (MDA8h O$_3$) and 24-h average PM$_{2.5}$. For both of these pollutants, National Ambient Air Quality Standards (NAAQS) have been established to protect against adverse human health effects (*4, 5*). Five cities in the continental United States—Atlanta, Chicago, Houston, Los Angeles, and New York—were chosen in this study because each experiences elevated ozone and PM$_{2.5}$ levels. Atlanta, Chicago, Los Angeles, and New York also have 24-h average PM$_{2.5}$ levels over the 35 μg m^{-3} NAAQS (http://www.epa.gov/oar/oaqps/greenbk/, last accessed 02/15/2008). Two years are chosen for this study: a "current" year, 2001, and "future", 2050. 2050 provides an opportunity to assess the combined effects of planned emission controls and climate change. Changes in sensitivities of MDA8h O$_3$ and 24-h average PM$_{2.5}$ to emissions are primarily due to planned emission changes between 2001 and 2050 as previous results suggest that the effects of emission controls are more significant than those of climate change alone (*2, 6*).

Methods

Quantifying sensitivities of air pollutant concentrations is done using EPA's Models-3 regional air quality system, applied as detailed elsewhere (*2, 6*), and described briefly here. The Fifth-Generation NCAR/Penn State Mesoscale meteorological Model (MM5) is used to downscale results (i.e., increase the spatial and temporal resolution over the chosen modeling domain) from NASA's Goddard Institute of Space Studies (GISS) (*7*) global climate model results for years 2001 and 2050 (*8, 9*). GISS results utilized are for the Intergovernmental Panel on Climate Change (IPCC) A1B scenario, which is generally viewed as a midrange case that

FIGURE 1. Daily sensitivities of MDA8h O_3 to anthropogenic NO_x emissions ($S_{MDA8h\ O_3,\ ANO_x}$) (in ppb, *Y*-axis) (based on a 1% change in emissions) versus MDA8h O_3 concentrations (in ppb, *X*-axis). Shown are the MDA8h O_3 and the corresponding (same day/time, same location) sensitivities in 2001 for city centers and regional maximum values (defined as the maximum over a 5 × 5 grid around the city) and in 2050 for city centers.

assumes a future world of rapid economic growth with a balance between fossil and nonfossil energy sources (*10*). Planned controls, e.g., the Clean Air Interstate Rule (CAIR) and others in the United States (*11*) as well as emission changes in Canada and Mexico (*12*) are used to forecast emissions to 2020. The Integrated Model to Assess the Global Environment (IMAGE) model (http://www.mnp.nl/image, last accessed 02/15/2008) is used to forecast emissions from 2020 to 2050. Emissions are processed by the Sparse Matrix Operator Kernel for Emissions (SMOKE) system version 2.1 (http://www.smoke-model.org/index.cfm, last accessed 02/ 15/2008). Anthropogenic SO_2 and NO_x emissions are projected to decrease 51% and 55%, respectively, between 2001 and 2050 over the simulation domain due to currently planned emission controls (Supporting Information (SI) Table S1) (*12*). Anthropogenic volatile organic compound (VOC) emissions are predicted to decrease about 38%, though total VOC emissions are projected to increase by about 2% as biogenic VOC emissions increase (SI Table S1). Ammonia (NH_3) emissions are predicted to increase by 7% due to growth in human activities (*6, 12*).

The Community Multiscale Air Quality Model (CMAQ) (*13*) version 4.3 with the SAPRC-99 (*14*) chemical mechanism and decoupled direct method 3D (DDM-3D) (*15, 16*) is used to simulate sensitivities of ozone and $PM_{2.5}$ to precursor emissions, including anthropogenic NO_x and VOC,

NH_3 and SO_2 emissions, over the domain covered by the continental United States and parts of Canada and Mexico in 2001 and 2050. A uniform grid of 36 × 36 km horizontal cells with 9 vertical layers is employed in the simulations (SI Figure S1). CMAQ with DDM-3D directly calculates the seminormalized first-order sensitivities of both gas- and condensed-phase pollutants to precursor emissions (*17, 18*), i.e., the seminormalized first-order sensitivity ($S_{i,j}$) of pollutant concentration i (C_i) to source emissions j (E_j) is determined as follows:

$$S_{i,j} = E_j \frac{\delta C_i}{\delta E_j} \tag{1}$$

The sensitivities, as presented here, have the same units as the corresponding pollutants. These sensitivities are local (accurate for small changes in emissions) and represent how pollutant concentrations respond to precursor emission changes as if the systems were linear. It is recognized that the system is not linear, but extensive testing suggests the first-order (linear) response is accurate up to emission changes of the order of 30% for ozone and 20−50% for $PM_{2.5}$ (depending on species) (*17–19*). Recognizing that changes by percent reductions in a source are more policy-relevant, here we show the daily sensitivities of ozone and $PM_{2.5}$ to 1% changes in emissions for the two years studied. Sensitivities

TABLE 1. Number of Days with Positive[a] and Negative[b] Sensitivities of MDA8h O_3 to Anthropogenic NO_x Emissions and MDA8h O_3 over Ozone Concentration of 85 ppb in 2001 and 2050 for the Five Cities

	2001 city center (+/−) (number of days[c] over 85 ppb)	2050 city center (+/−) (number of days over 85 ppb)	2001 (+/−) regional maximum (number of days over 85 ppb)
Atlanta	144/215 (66)	276/83 (0)	224/135 (87)
Chicago	31/328 (19)	131/218 (6)	188/171 (28)
Houston	117/242 (35)	294/65 (3)	262/97 (58)
Los Angeles	87/272 (9)	285/74 (1)	125/234 (56)
New York	3/356 (0)	79/280 (0)	173/186 (31)

[a] Positive sensitivity (+): Reductions in anthropogenic NO_x emissions decrease MDA8h ozone levels. [b] Negative sensitivity (−): Reductions in anthropogenic NO_x emissions increase MDA8h ozone levels. [c] The first seven days of each year are excluded to minimize the impacts of initial concentrations, leaving 359 days for analysis.

of ozone and $PM_{2.5}$ are examined for the grid over the city center where population densities are typically highest, and also at the location of the regional ozone maximum (i.e., maximum values among five × five grid cells around the city center, SI Figure S1). While the ozone response at the city center has increased utility in health-based analyses (city-center monitors are often used in health effects studies, and generally are associated with high population densities), the regional maximum is used in design of strategies to meet the ozone NAAQS.

Results and Discussion

Daily Linked Responses of Daily Maximum 8-h Average Ozone and 24-h Average $PM_{2.5}$ to Anthropogenic NO_x and VOC Emissions. The response (or sensitivity, S) of the MDA8h O_3 to anthropogenic NO_x emissions ($S_{MDA8h\ O_3,\ ANO_x}$) is typically correlated with the corresponding MDA8h O_3 levels (Figure 1; SI Table S2 provides correlation statistics) when viewed on a daily basis for the years studied. Reductions in anthropogenic NO_x emissions are usually effective in decreasing MDA8h O_3 concentrations on days of higher O_3, both at the city center as well as at the regional maximum (Figure 1). On the other hand, reductions in anthropogenic NO_x emissions are expected to increase MDA8h O_3 concentrations on days less conducive to ozone formation, a response found more at the city center (where, depending on the city, 215−356 days have this adverse response, Table 1) than for the regional daily maximum (where 97−234 days have a negative sensitivity). The forecast 55% reduction in domain-wide anthropogenic NO_x emissions between 2001 and 2050 is shown to make the formation of moderate-level ozone more NO_x-limited and the sensitivities of MDA8h O_3 to anthropogenic NO_x more positive in 2050 as compared with 2001 (Figure 1). Further, the highest MDA8h O_3 levels are reduced between 2001 and 2050, though levels are simulated to increase on low-ozone days (Figure 1). MDA8h O_3 levels and sensitivities of MDA8h O_3 to anthropogenic NO_x emissions are predicted to have a higher correlation in 2050 ($0.53 < r^2 < 0.81$, depending upon city) than 2001 $0.0 < r^2 < 0.77$ (Figure 1 and SI Table S2), and the slopes are typically higher as well. Slopes in 2001 range from 0.0 to 0.006 (ppb/%)/ppb, and increase to 0.005 to 0.010 (ppb/%)/ppb in 2050, showing that NO_x controls are more efficient in reducing MDA8h O_3 concentrations in 2050 than 2001 for the five cities (Table 1) and there are fewer cases where ozone has a negative response. Based on a 1% change in anthropogenic NO_x emissions in 2001, sensitivities of MDA8h O_3 to anthropogenic NO_x emissions are simulated to vary from about −0.3 to +0.4 ppb depending on prevailing NO_x abundance in the five cities (Figures 1 and 2). Sensitivities of MDA8h O_3 to VOC are typically positive (though often small), and negatively correlated with NO_x sensitivities (SI Figure S2). VOC sensitivities are greater in 2001 versus 2050. While reductions in anthropogenic VOC emissions always
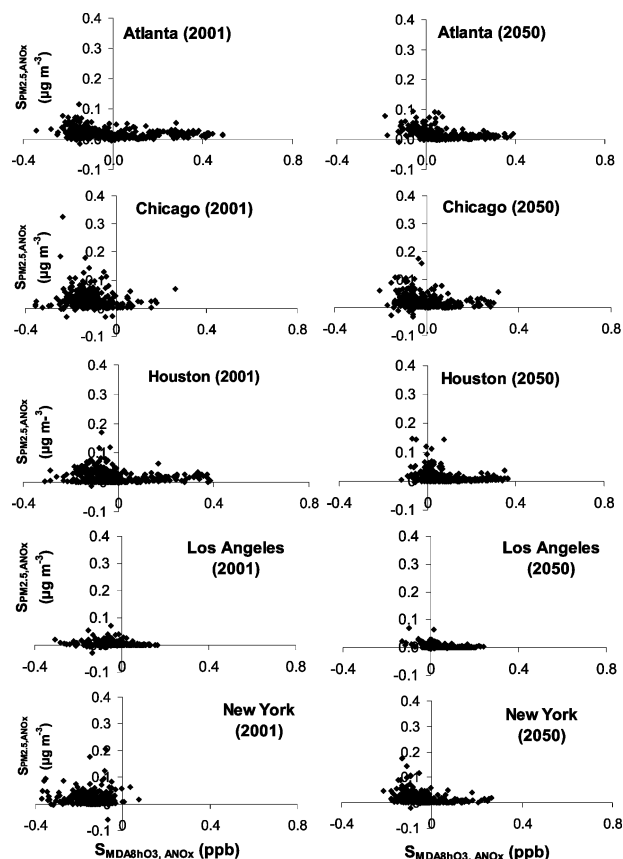
FIGURE 2. Daily sensitivities of 24-h $PM_{2.5}$ ($S_{PM2.5,\ ANO_x}$, in $\mu g\ m^{-3}$, Y-axis) and MDA8h O_3 to anthropogenic NO_x emissions ($S_{MDA8h\ O_3,\ ANO_x}$, in ppb, X-axis) in 2001 and 2050 for city centers (each shown as response to a 1% change in anthropogenic NO_x emissions).

decrease MDA8h O_3 levels in 2001 there are a few days where there is a slightly negative response in 2050 (Figure 1 and SI Figure S2). For regional maximum MDA8h O_3 in 2001, NO_x-sensitive environments become "NO_x-starved" and the correlation between the sensitivity of MDA8h O_3 to NO_x and MDA8h O_3 concentrations is stronger ($0.1 < r^2 < 0.84$) and slope also increases for four of the five cities as compared with city-center MDA8h O_3 (SI Table S2).
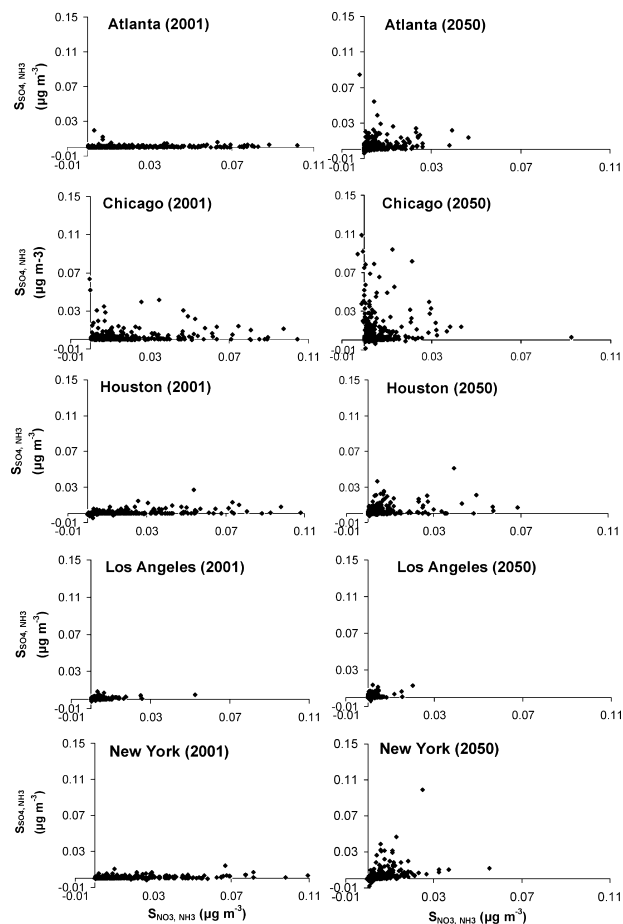
Sensitivities of 24-h average $PM_{2.5}$ to anthropogenic NO_x emissions ($S_{PM2.5,\ ANO_x}$) are predicted to range from about 0 to 0.1 $\mu g\ m^{-3}$ in 2001 and 2050 based on 1% change in anthropogenic NO_x emissions. Reductions in anthropogenic NO_x cause decreases in nitrate but slight increases in sulfate formation (2). The net effects show a positive $S_{PM2.5,\ ANO_x}$ in 2001 and 2050 (Figure 2). This suggests that reductions in anthropogenic NO_x emissions are expected to continue to
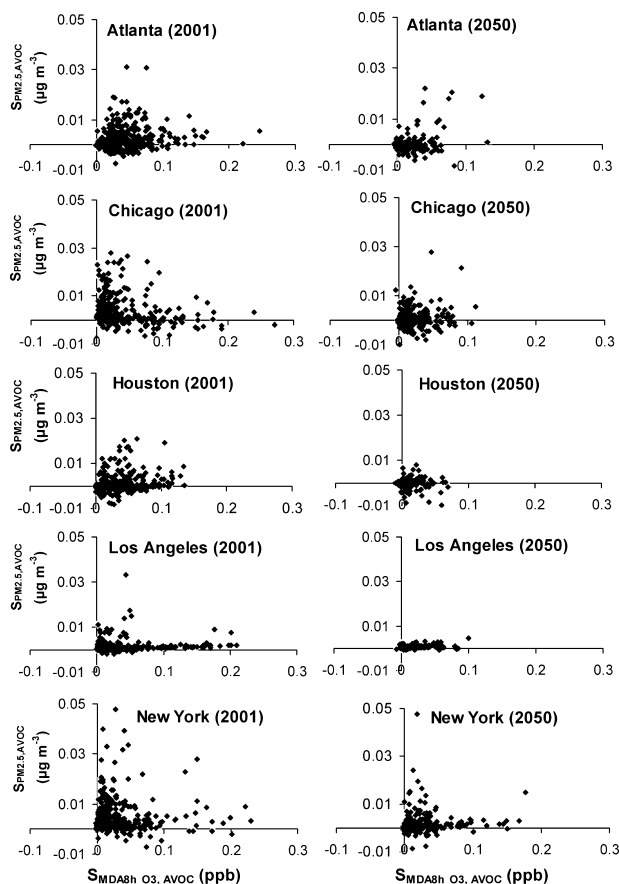
FIGURE 3. Daily sensitivities of 24-h PM$_{2.5}$ (S$_{PM2.5, AVOC}$, in μg m^{-3}, Y-axis) and MDA8h O$_3$ to anthropogenic VOC emissions (S$_{MDA8h\ O_3,\ AVOC}$, in ppb, X-axis) in 2001 and 2050 for city centers (each shown as response to a 1% change in anthropogenic VOC emissions).

FIGURE 4. Daily sensitivities of sulfate (S$_{SO_4,\ NH_3}$, in μg m^{-3}, Y-axis) and nitrate (S$_{NO_3,\ NH_3}$, in μg m^{-3}, X-axis) to NH$_3$ emissions in 2001 and 2050 for city centers (each shown as response to a 1% change in NH$_3$ emissions).

be effective in deceasing 24-h PM$_{2.5}$ (Figure 2), and such controls will tend to be more effective and positive for reducing ozone in the future.

Ozone and PM$_{2.5}$ responses to VOC controls are likewise linked. Sensitivities of MDA8h O$_3$ concentrations to anthropogenic VOC emissions (S$_{MDA8h\ O_3,\ AVOC}$) range from about 0 to 0.2 ppb while sensitivities of 24-h average PM$_{2.5}$ concentrations to anthropogenic VOC emissions (S$_{PM2.5,\ AVOC}$) are simulated to vary from −0.005 to +0.02 μg m^{-3} based on a 1% change in anthropogenic VOC emissions in 2001 (Figure 3). Positive sensitivities of MDA8h O$_3$ to anthropogenic VOC emissions imply that reductions in anthropogenic VOC emissions are effective in decreasing MDA8h O$_3$. On the other hand, there are a few cases where sensitivities of 24-h PM$_{2.5}$ to anthropogenic VOC emissions suggest that reductions in anthropogenic VOC emissions may slightly increase 24-h PM$_{2.5}$ levels. This is attributed to interdependencies among anthropogenic VOCs, radicals, SO$_2$, and NO$_x$ levels in the ambient air (18). Reductions in anthropogenic VOC emissions decrease secondary organic aerosol (SOA) formation but can increase OH radical levels, more rapidly oxidizing SO$_2$ and NO$_x$ which can increase PM$_{2.5}$ concentrations. In 2050, sensitivities of MDA8h O$_3$ and 24-h average PM$_{2.5}$ to anthropogenic VOC emissions are predicted to decrease mainly due to planned reductions in anthropogenic VOC emissions between 2001 and 2050 (Figure 3). It is important to note that current air quality models do not fully capture SOA formation (20), and the actual PM$_{2.5}$ sensitivities are likely to be more positive than simulated, though they highlight the linkage between the responses of O$_3$ and PM$_{2.5}$ to anthropogenic VOC emissions.

Linked Responses of Sulfate and Nitrate to NH$_3$, SO$_2$, and NO$_x$ Emissions. Sensitivities of nitrate to NH$_3$ emissions (S$_{NO_3,\ NH_3}$) (up to about 0.1 μg m^{-3} based on 1% change in NH$_3$ emissions) are found to be much higher than sensitivities of sulfate to NH$_3$ emissions (S$_{SO_4,\ NH_3}$) (up to about 0.02 μg m^{-3} based on 1% change in NH$_3$ emissions, which is about 1/5 of the nitrate responses) in 2001 in the five cities (Figure 4). High sensitivities of nitrate to NH$_3$ emissions are due to the thermodynamic equilibrium among sulfate, nitrate, and ammonium. Formation of NH$_4$NO$_3$ is limited by availability of ammonium (NH$_3$) after (NH$_4$)$_2$SO$_4$ is formed. This is particularly true in areas with high NO$_x$ and SO$_2$ emissions. In 2050, higher temperatures and humidity increase hydroxyl radicals and induce more rapid oxidation of SO$_2$ and NO$_x$. Also, pH-dependent aqueous phase oxidation of sulfate becomes more important. However, higher temperatures also increase gas-phase partitioning of semivolatile PM$_{2.5}$ compounds, such as NH$_4$NO$_3$. Overall, lower anthropogenic SO$_2$ and NO$_x$ emissions, and higher ammonia emissions and temperatures cause NH$_4$NO$_3$ formation to become less ammonia-sensitive in 2050. The increased importance of aqueous-phase oxidation of SO$_2$ causes (NH$_4$)$_2$SO$_4$ formation to become more ammonia-sensitive even though SO$_2$ emissions are predicted to decrease in 2050 due to planned emission controls. Overall, the sensitivities of (NH$_4$)$_2$SO$_4$ to NH$_3$ increase, a finding that is opposite of the one for NH$_4$NO$_3$.

Sensitivities of sulfate to SO$_2$ emissions (S$_{SO_4,\ SO_2}$) and nitrate to anthropogenic NO$_x$ emissions (S$_{NO_3,\ ANO_x}$) are simulated to be mainly positive in 2001 and 2050 (Figure 5 and SI Figure S3). Reductions in SO$_2$ and anthropogenic NO$_x$ emissions, respectively, are predicted to decrease gas- and
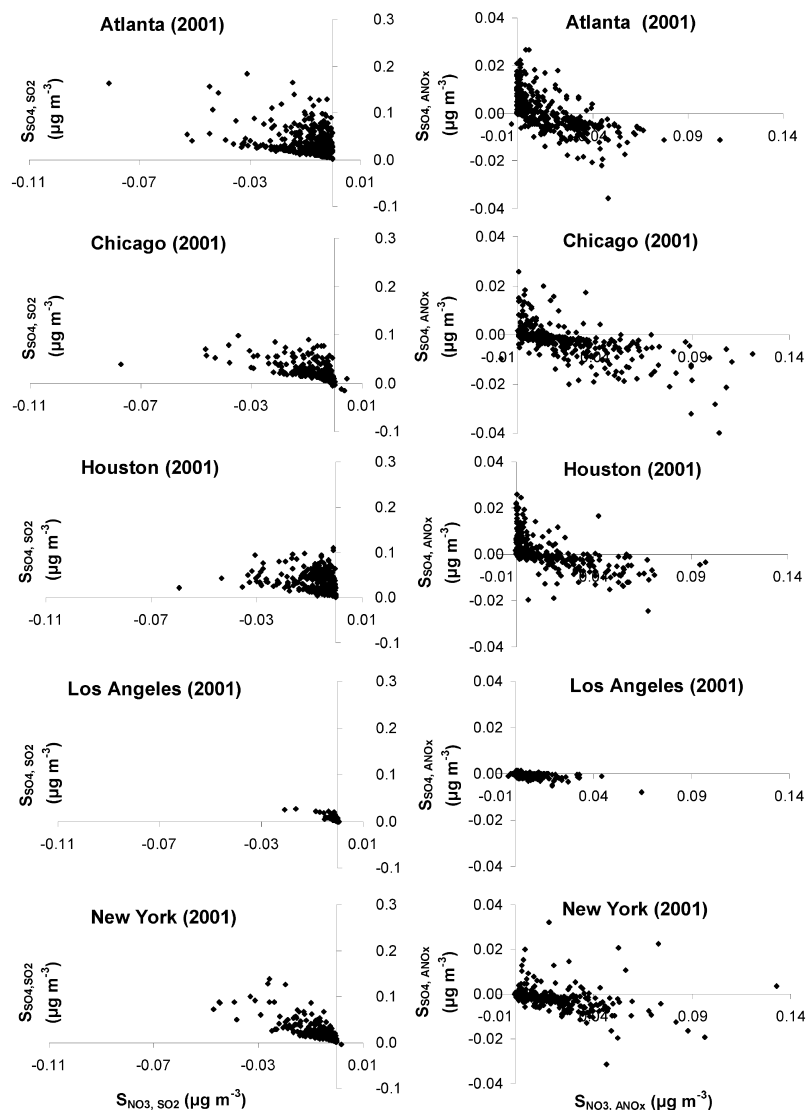
FIGURE 5. Daily sensitivities of sulfate and nitrate (in μg m^{-3}, X-axis) to 1% changes in SO$_2$ emissions (S$_{SO_4, SO_2}$ and S$_{NO_3, SO_2}$, in μg m^{-3}, left column) and anthropogenic NO$_x$ emissions (S$_{SO_4, ANO_x}$ and S$_{NO_3, ANO_x}$, in μg m^{-3}, right column) in 2001 for city centers.

aqueous-phase sulfate and nitrate, and lead to less condensable (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ formation in 2050. On the other hand, competition for ammonia/ammonium between nitrate and sulfate causes sensitivities of nitrate to SO$_2$ emissions (S$_{NO_3, SO_2}$) to be negative and, therefore, reductions in SO$_2$ emission are simulated to increase nitrate formation. Nevertheless, when lowering NO$_x$ emissions reduce oxidant levels (e.g., OH, H$_2$O$_2$, O$_3$, etc.), sulfate formation can decrease (i.e., when sensitivities of sulfate to anthropogenic NO$_x$ emissions (S$_{SO_4, ANO_x}$) are positive). Reductions in SO$_2$ and anthropogenic NO$_x$ emissions are simulated to lead to similar decreases in annual 24-h average PM$_{2.5}$ concentrations (SI Table S3). Both future (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ are found to be less sensitive to SO$_2$ and anthropogenic NO$_x$ emissions due to controls (Figure 5 and SI Figure S3).

Current Annual Average Responses. While viewing the daily linked sensitivities of ozone and PM$_{2.5}$ to emissions provides a rapid assessment of the complexities in the effects of controls, some health effects are linked to more chronic exposures to these pollutants, and many areas experience annual PM$_{2.5}$ levels above the NAAQS. Further, acute responses to daily maximum ozone levels are found as well (21). While the sensitivity of the fourth highest, regional maximum, MDA8h O$_3$ to NO$_x$ is positive for all the cities except Los Angeles (SI Table S3), the annual average of the

NO$_x$ sensitivities of the MDA8h O$_3$ is negative for four of the five cities (from −0.15 to −0.01 ppb/%; Atlanta being the exception). Further, the annual average ozone response to NO$_x$ is negative at all locations (from −0.11 to −0.05 ppb/%). All of the annual average ozone metrics are found to respond positively to VOC controls. Annual average PM$_{2.5}$ will be reduced by SO$_2$ and NO$_x$ reductions, with sensitivities of 0.0−0.04 μg m^{-3}/% for SO$_2$ reductions and 0.01−0.03 μg m^{-3}/% for NO$_x$ reductions (SI Table S3).

Consideration of responses of ozone and PM$_{2.5}$ to emission changes shows the complexities in choosing optimum strategies to address air quality problems. While NO$_x$ control is shown to reduce ozone on days with the most elevated ozone levels, it can raise ozone on others. The response of ozone in the city center and the location of the regional maximum are similar in three cities, though not in New York and Chicago. Both ozone and PM$_{2.5}$ are reduced in response to VOC controls, but not in response to NO$_x$. There is an inverse relationship between how sulfate and nitrate respond to both SO$_2$ and NO$_x$ controls. Further, the response of the annual averages is quite different from that of peak daily levels for both PM$_{2.5}$ and ozone, so health effects associated with acute exposures will respond differently than health effects associated with chronic exposures. This also impacts

formulating strategies to meet the various NAAQS, including daily maximum ozone and PM$_{2.5}$, as well as the annual average PM$_{2.5}$.

Acknowledgments

Supporting Information Available

Additional figures and data tables. This material is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited

(1) Lamarque, J. F.; Kiehl, J. T.; Hess, P. G.; Collins, W. D.; Emmons, L. K.; Ginoux, P.; Luo, C.; Tie, X. X. Response of a coupled chemistry-climate model to changes in aerosol emissions: Global impact on the hydrological cycle and the tropospheric burdens of OH, ozone, and NOx. *Geophys. Res. Lett.* **2005**, 32.

(2) Liao, K. J.; Tagaris, E.; Manomaiphiboon, K.; Napelenok, S. L.; Woo, J. H.; He, S.; Amar, P.; Russell, A. G. Sensitivities of ozone and fine particulate matter formation to emissions under the impact of potential future climate change. *Environ. Sci. Technol.* **2007**, *41*, 8355–8361.

(3) Unger, N.; Shindell, D. T.; Koch, D. M.; Streets, D. G. Cross influences of ozone and sulfate precursor emissions changes on air quality and climate. *Proc. Natl. Acad. Sci., U.S.A.* **2006**, *103*, 4377–4380.

(4) Burnett, R. T.; Cakmak, S.; Brook, J. R.; Krewski, D. The role of particulate size and chemistry in the association between summertime ambient air pollution and hospitalization for cardiorespiratory diseases. *Environ. Health Perspect.* **1997**, *105*, 614–620.

(5) Lippmann, M. Health-Effects of Tropospheric Ozone - Review of Recent Research Findings and Their Implications to Ambient Air-Quality Standards. *J. Exposure Anal. Environ. Epidemiol.* **1993**, *3*, 103–129.

(6) Tagaris, E.; Manomaiphiboon, K.; Liao, K. J.; Leung, L. R.; Woo, J. H.; He, S.; Amar, P.; Russell, A. G. Impacts of Global Climate Change and Emissions on Regional Ozone and Fine Particulate Matter Concentrations over North America. *J. Geophys. Res., Atmos.* **2007**, 112.

(7) Rind, D.; Lerner, J.; Shah, K.; Suozzo, R. Use of on-line tracers as a diagnostic tool in general circulation model development 2. Transport between the troposphere and stratosphere. *J. Geophys. Res., Atmos.* **1999**, *104*, 9151–9167.

(8) Mickley, L. J.; Jacob, D. J.; Field, B. D.; Rind, D. Effects of future climate change on regional air pollution episodes in the United States. *Geophys. Res. Lett.* **2004**, 31.

(9) Leung, L. R.; Gustafson, W. I. Potential regional climate change and implications to US air quality. *Geophys. Res. Lett.* **2005**, 32.

(10) IPCC. *Climate Change 2001: The Science of Climate Change*; Intergovernmental Panel on Climate Change, 2001.

(11) Houyoux, M. *CAIR Emissions Inventory Overview*; U.S. EPA: Washington, DC, 2004.

(12) Woo, J. H.; He, S. P.; Amar, P.; Tagaris, E.; Manomaiphiboon, K.; Liao, K. J.; Russell, A. G. Development of Mid-Century Anthropogenic Emissions Inventory in Support of Regional Air Quality Modeling under Influence of Climate Change. *J. AWMA* (accepted).

(13) Byun, D. W.; Schere, K. L. Review of the governing equations, computational algorithms, and other components of the Models-3 Community Multscale Air Quality (CMAQ) modeling system. *Appl. Mech. Rev.* **2006**, 51–77.

(14) Carter, W. P. L. *Documentation of the SAPRC-99 Chemical Mechanism for VOC Reactivity Assessment*; Final Report to California Air Resources Board, Contracts 92-329 and 95-308, 2000.

(15) Dunker, A. M. The Decoupled Direct Method for Calculating Sensitivity Coefficients in Chemical-Kinetics. *J. Chem. Phys.* **1984**, *81*, 2385–2393.

(16) Dunker, A. M.; Yarwood, G.; Ortmann, J. P.; Wilson, G. M. The decoupled direct method for sensitivity analysis in a three-dimensional air quality model - Implementation, accuracy, and efficiency. *Environ. Sci. Technol.* **2002**, *36*, 2965–2976.

(17) Cohan, D. S.; Hakami, A.; Hu, Y. T.; Russell, A. G. Nonlinear response of ozone to emissions: Source apportionment and sensitivity analysis. *Environ. Sci. Technol.* **2005**, *39*, 6739–6748.

(18) Napelenok, S. L.; Cohan, D. S.; Hu, Y.; Russell, A. G. Decoupled direct 3D sensitivity analysis for particulate matter (DDM-3D/PM). *Atmos. Environ.* **2006**, *40*, 6112–6121.

(19) Hakami, A.; Odman, M. T.; Russell, A. G. Nonlinearity in atmospheric response: A direct sensitivity analysis approach. *J. Geophys. Res., Atmos.* **2004**, 109.

(20) Morris, R. E.; Koo, B.; Guenther, A.; Yarwood, G.; McNally, D.; Tesche, T. W.; Tonnesen, G.; Boylan, J.; Brewer, P. Model sensitivity evaluation for organic carbon using two multi-pollutant air quality models that simulate regional haze in the southeastern United States. *Atmos. Environ.* **2006**, *40*, 4960–4972.

(21) Bell, M. L.; McDermott, A.; Zeger, S. L.; Samet, J. M.; Dominici, F. Ozone and short-term mortality in 95 US urban communities, 1987−2000. *JAMA, J. Am. Med. Assoc.* **2004**, *292*, 2372–2378.

ES7028685